FINE PARTICLES

Aerosol Generation, Measurement, Sampling, and Analysis

Edited by
BENJAMIN Y. H. LIU

ABSTRACT

GENERATION OF MONODISPERSE SUBMICRON AEROSOLS BY ABLATION FROM TRANSPIRATION COOLED POROUSE MATRICES

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Academic Press, Inc.
New York, N. Y. 10003

GENERATION OF MONODISPERSE SUBMICRON AEROSOLS BY ABLATION FROM TRANSPIRATION-COOLED POROUS MATRICES°°

by

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ABSTRACT

In this paper the generation is described of monodisperse submicron particles of controlled size by ablation from transpiration-cooled porous matrices.

A new experimental apparatus is illustrated which has been realized in the Istituto di Fisica Tecnica - Politecnico di Torino - Italy, and which is based on previous works in cooperation with the University of Minnesota, USA and the Sonderforschungsbereich 80, Universität Karlsruhe, West Germany.

In the new version of the apparatus an ewternal heat source (a plasma jet) heats a porous matrix up to ablation temperatures and particles are generated by ablation of the porous material itself. The temperature of the ablated surface of the porous matrix is controlled by a flow of gas transpiring through the matrix, which also quenches and dilutes the particles formed in the ablation process. Particles can be generated from any porous material of sufficiently fine structure and uniform porosity, in particular from graphite, tungsten, ceramics, etc.

(°°) This work has been sponsored by the Italian National Research Council, Centro Nazionale delle Ricerche.

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The experimental apparatus is described as well as the particle measuring techniques, based on the utilization of a calibrated T.S.I. Model 3030 Whitby Electrical Aerosol Analyzer.
Experimental results are given referring to the generation of submicron graphite particles. The measured number, surface, and volume distributions of these particles are given, together with the logarithmic standard deviation σg. The results show that, for the generated particles $1.13 < \sigma g < 1.32$.

GENERATION OF MONODISPERSE SUBMICRON AEROSOLS BY ABLATION FROM TRANSPIRATION-COOLED POROUS MATRICES °

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INTRODUCTION

This report describes a new approach for the generation of monodisperse aerosols which are presently of interest for all kinds of medical and air pollution research, including evaluating aerosol sampling and measuring equipment, air cleaner evaluation, inhalation studies, and for interrelated studies. Although numerous research-type aerosol generators utilising electric arcs have been developed or proposed for generating monodisperse aerosols ranging from 0.02 to 20 μm, there is at present no reliable method available for generating ultrafine, monodisperse aerosols for a wide spectrum of materials, and in particular of refractory materials, based on the utilisation of electric arcs.

Aerosol generation by means of electric arcs between solid electrodes relies on the high temperature of the attachment spots at the electrodes which causes an evaporation of some electrode material. This metal vapor, if cooled rapidly, condenses into an aerosol. The properties of the aerosol particles, including their size, depend on a number of parameters, particularly on the temperature distribution at

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and in the vicinity of the arc attachment spot. It has not been possible to generate particles with a nearly uniform size or monodisperse particles under these conditions because of the temperature variations and the varying conditions at the location at which individual particles are generated. The time needed to collect a sufficient number of particles is large compared to the characteristic time during which changes on the surface occur, i.e., the relevant parameters cannot be held constant during this period. The main reason for these varying conditions lies in the nature of the attachment spot of the arc which moves more or less quickly over the electrodes, even if current and voltage do not fluctuate appreciably.

But these variations, in addition to the explosive character of the vaporization process, change the conditions for the aerosol production so rapidly that for any useful length of time the produced aerosol has a wide distribution of its particle size.

Various experimental configurations have been suggested for the production of aerosols with these techniques (Chace et als[1], Reichelt[2], Holmgren et als[3], Kuhn[4]) employing both high and low intensity arcs. Solid electrodes have been used by all investigators. The electrodes have frequently been cooled by a conventional water cooling system, especially in conjunction with high intensity arcs. With low intensity arcs coagulated and chainlike particles are produced. The size distribution of these particles covers a wide range, from approximately 100 to 1000 Å (Reichelt[2]) and is poorly controlled. Better results have been obtained by using high intensity arcs (Holmgren et als[3]). The feed material is incorporated in the body of the anode which consumes a large fraction of the arc power. In most cases the solid electrodes are water-cooled and dilution gas can be blown through the arc. By varying the cooling rate of the electrodes and the dilution flow rate through the plasma in the region of maximum condensation, the particle size may to a certain degree be controlled. Monodisperse spheroidal shaped particles with a diameter ranging from approximately 100 to 1000 Å have been obtained.

The results of the above-discussed investigations indicate that electric arcs are a poor source for generating aerosols. It seems to be impossible to obtain small monodisperse particles of uniform size because coagulation occurs as soon as the particles are produced. In addition, the particle generation process is characterized by strong non-uniformities which are difficult to control in conventional arc arrangements with solid electrodes.

During the past two years, a new method for the generation of ultrafine, monodisperse aerosol has been successfully explored (Boffa et als[5]). This method makes use of a transpiration-cooled anode in a high intensity arc. In this situation, the aerosol is generated continuously rather than in an explosive manner at the steady attachment spot which is the hottest part of the anode surface. The transpiring gas passing through the anode provides quenching of the vapor and an immediate dilution of the generated particles in the vicinity of the anode surface avoiding in this way coagulation and formation of agglomerations.

The method as such has been very successful, but the particle generator itself is a rather complicated device which requires skilled personnel for its proper operation. The knowledge acquired during previous work has been, however, instrumental for the success of the new approach described in this report.

In the new approach here described the arc plasma source is entirely separated from the body from which the aerosol is generated; but the basic features for the aerosol production process inherent to the previously-developed device are retained. Instead of a transpiration cooled anode which is an integral part of the previously-developed plasma torch, a porous, transpiration-cooled body will be utilized heated by the plasma jet emanating from a conventional arc plasma torch. Since transpiration cooling fullfills two tasks simultaneously; namely, quenching of the generated vapor and dilution of the particles formed in the quenching process, utilization of this mechanism is considered as the basic principle which also governs this approach.

The separation of the heat source from the particle providing surface has at least two important advantages over the previous method:

I) The resulting particle generator is simple and straightforward to operate, and easily movable, as required by the experiments which will be performed in this research program.

II) The useful parameter range for operation of this device and the associated flexibility is substantially enhanced.

EXPERIMENTAL APPARATUS

The experimental apparatus consists essentially of a commercial power supply and control unit, which enable the operation of a plasma torch, which in turn represents the heat source, necessary for the operation of the aerosol generator.
The plasma torch and the aerosol generator are schematically represented in fig. 1.
The plasma torch consists essentially of a water cooled, thoriated tungsten cathode, with a conical tip, and a nozzle shaped water cooled copper anode, coaxial with the cathode. Argon is injected tangentially at the base of the catode, thus providing a swirl flow around the cathode tip, which stabilizes the arc. This gas, passing through the arc, emerges from the anode nozzle in the form of a highly energized plasma jet.
The cooling of anode and cathode is provided by a water cooling system, as indicated in fig. 2.
For an overall energy balance the cooling water flow is measured by means of flowrators, and the temperature increase of the anode and cathode cooling water is measured by means of four thermocouples located in the water circuit immediately upstream and downstream from the electrodes.
In this way the energy losses to the electrodes, that is the heat transferred from the arc to the anode and cathode, can be determined. The power in the emerging plasma jet can be

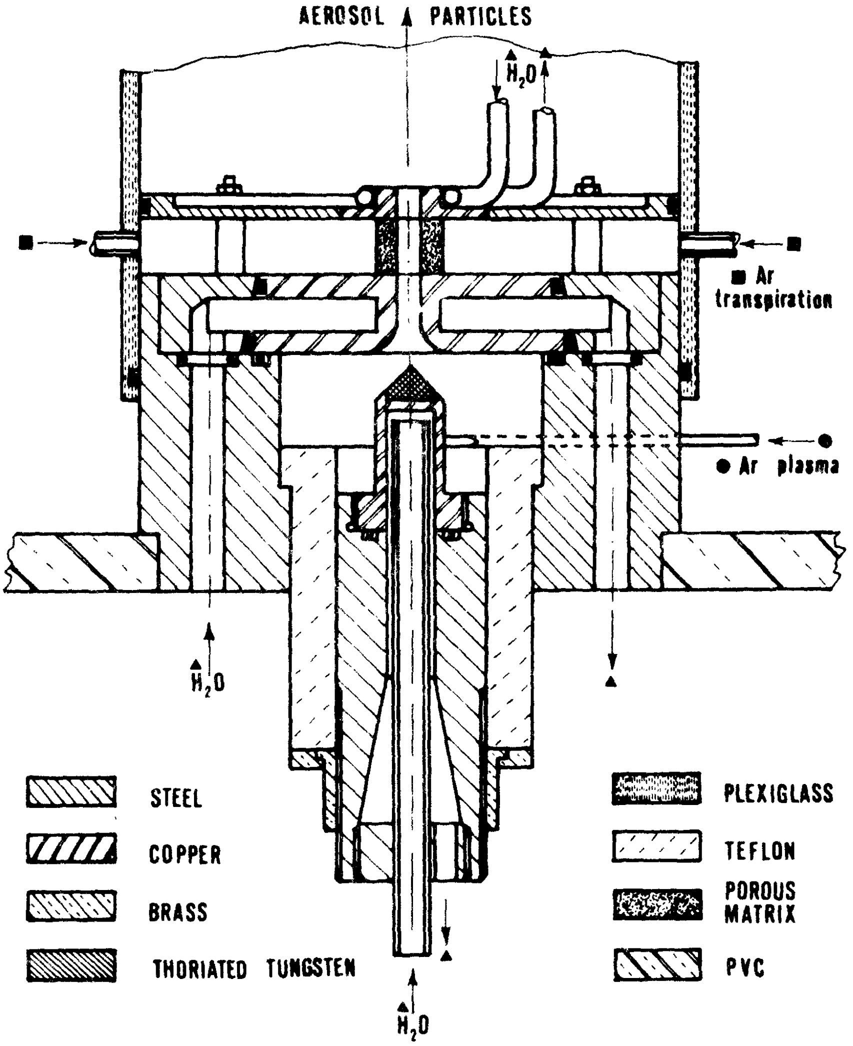

Fig. 1 Schematic of the plasma torch with aerosol generator.

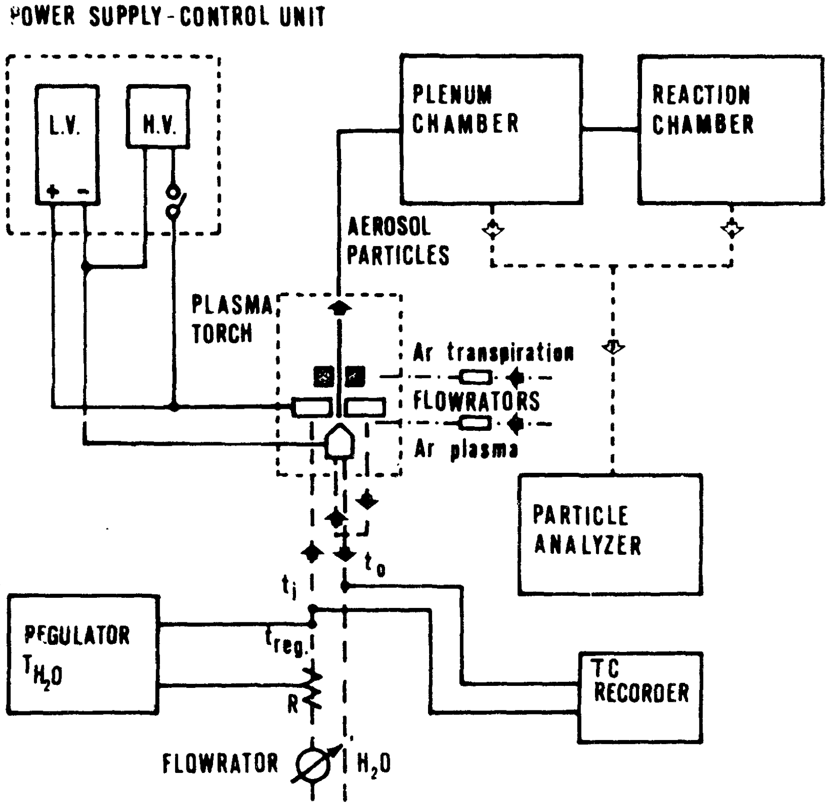

Fig. 2 Schematic of the experimental set-up.

calculated as the difference between the electrical power input and the power losses to the electrodes, carried away by the cooling water.

As before mentioned, the plasma jet emerging from the anode nozzle enters the aerosol generator located on top of the plasma torch and coaxial with it.

The aerosol generator itself consists essentially of a transpiration cooled porous carbon nozzle, concentrical with the plasma jet, inbedded in a steel and plexiglass structure which supports the porous nozzle and acts as plenum chamber feeding the cooling gas to the nozzle itself.

The cooling gas transpires through the nozzle and emerges into the plasma jet, diluting the aerosol particles, which are continuously produced by ablation from the porous carbon nozzle inside surface facing the plasma.

A cylindrical plexiglass window surrounds the plenum chamber and seals it, by means of "O" rings. Through this window pyrometric readings of the outer surface temperature of the porous nozzle are possible. The plenum chamber is equipped with an inlet for the cooling gas and with a pressure tap. The steel portion of the structure supporting the porous nozzle is water cooled.

The aerosol emerging from the generator, in suspension in the argon gas, is collected into a 2 m3 collection chamber, where it can be additionally diluted.

A block diagram of the experimental setup is shown in fig. 2.

The aerosol size distribution measuring system consists mainly of a calibrated T.S.I. Model 3030 Whitby Electrical Aerosol Analizer described elsewhere (Liu et als$^{6)}$).

EXPERIMENTAL RESULTS AND CONCLUSIONS

The results referring to carbon particles are summarized in fig. 3 to 7.

In fig. 3 the normalized number distributions of the produced aerosols are depicted, corresponding to different values of the argon flowrate transpiring through the ablat-

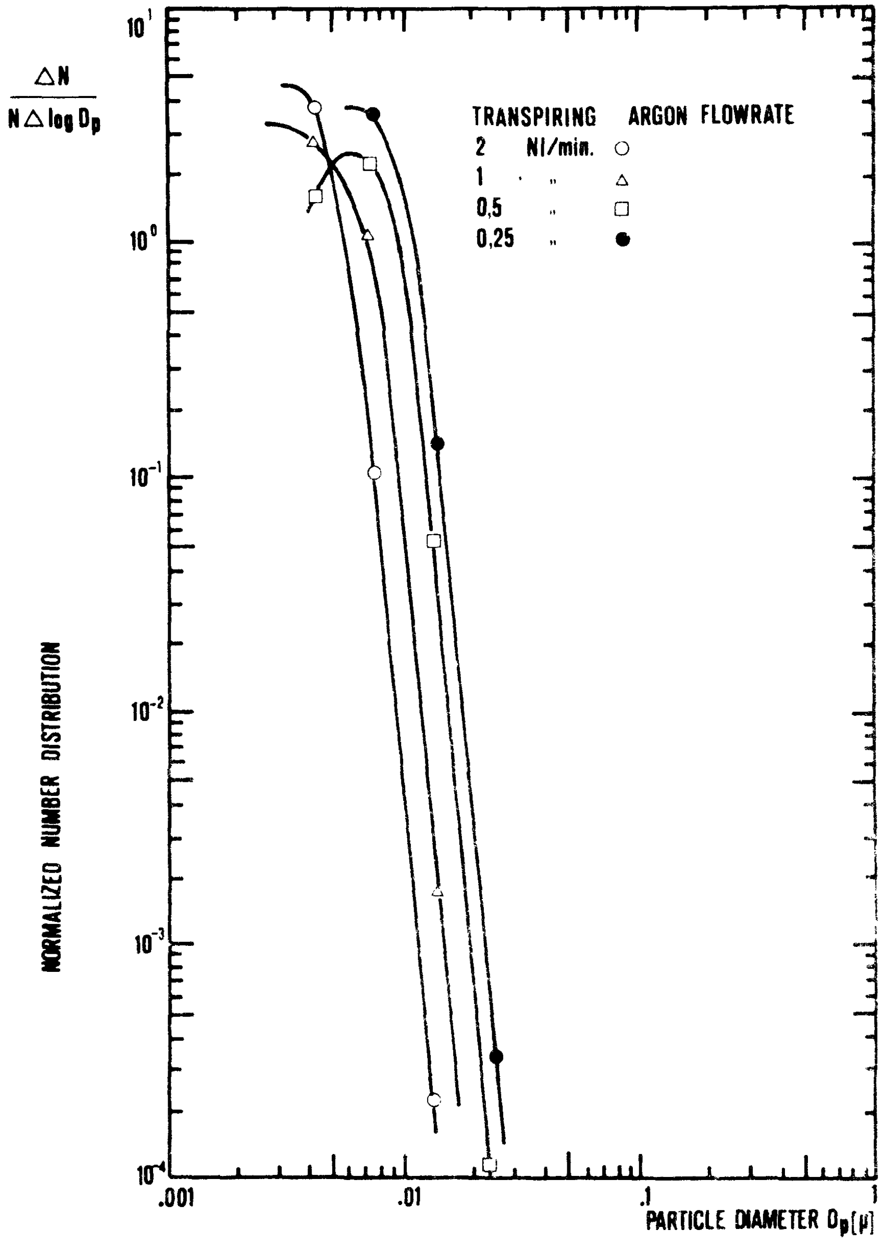

Fig. 3 Normalized number distributions of the generated aerosols.

ing porous matrix. Figures 4 and 5 show the corresponding normalized surface and volume distributions.

All the data are taken with an arc current of 100 Amp.

The results show that, for a given arc current value, the mean diameter of the generated aerosol decreases with increasing of the argon flowrate transpiring through the ablating matrix.

This finding can be qualitatively understood considering the influence of the transpiring gas flow-rate on the temperature of the ablating surface of the porous nozzle. By increasing the transpiring mass flow-rate, the temperature of the ablating surface and the associated rate of ablation decreases, yielding smaller particle densities, lower coagulation rates and, therefore, smaller particle diameters.

At the same time, an increase in transpiring mass flow-rate enhances the dilution of the ablated particles in front of the surface, which decreases the coagulation rate and, consequently, the particle size also.

In fig. 6 the logaritmic standard deviation is depicted as a function of the transpiring argon flowrate for an arc current of 100 Amp.

It can be seen that for the generated particles $1.13 < \sigma g < 1.32$.

The logarithmic standard deviation, characterizing the ablated aerosol size distribution, is a decreasing function of the transpiring mass flow-rate. It is speculated that the gas transpiring through the ablating surface has a quenching effect on the particles immediately after they are produced, preventing them from further coagulation.

The higher the gas flow-rate, the smaller the number of particles which can undergo coagulation over an extended time period and randomly increase their size, resulting in a wider size distribution. On the basis of these results, it is obvious that, keeping the arc current at the fixed value of 100 A, the logarithmic standard deviation decreases with increasing transpiring argon flowrate, as shown in fig. 6.

In fig. 7 the generated aerosol particles total number density is plotted as a function of the transpiring argon flowrate for an arc current of 100 Amp,

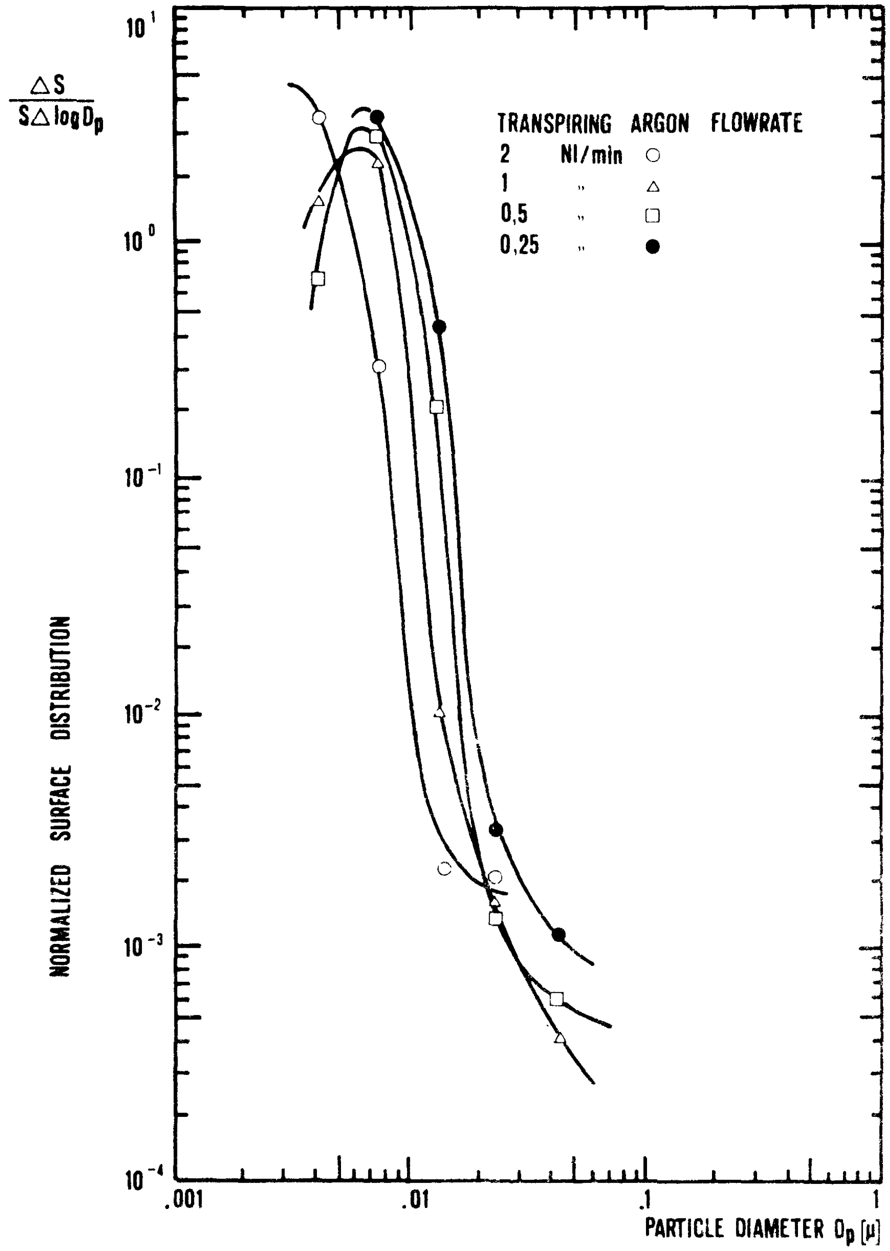

Fig.4 Normalized surface distributions of the generated aerosols.

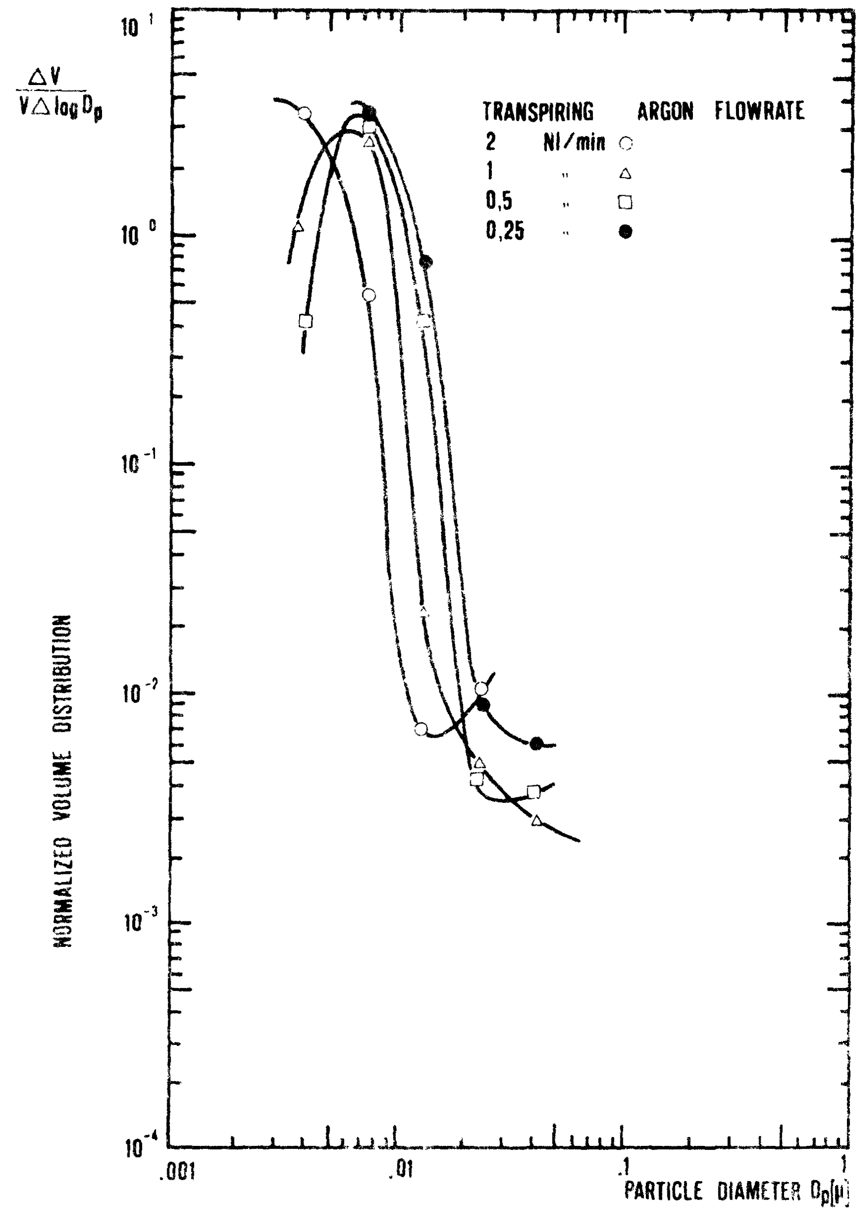

Fig. 5 Normalized volume distributions of the generated aerosols.

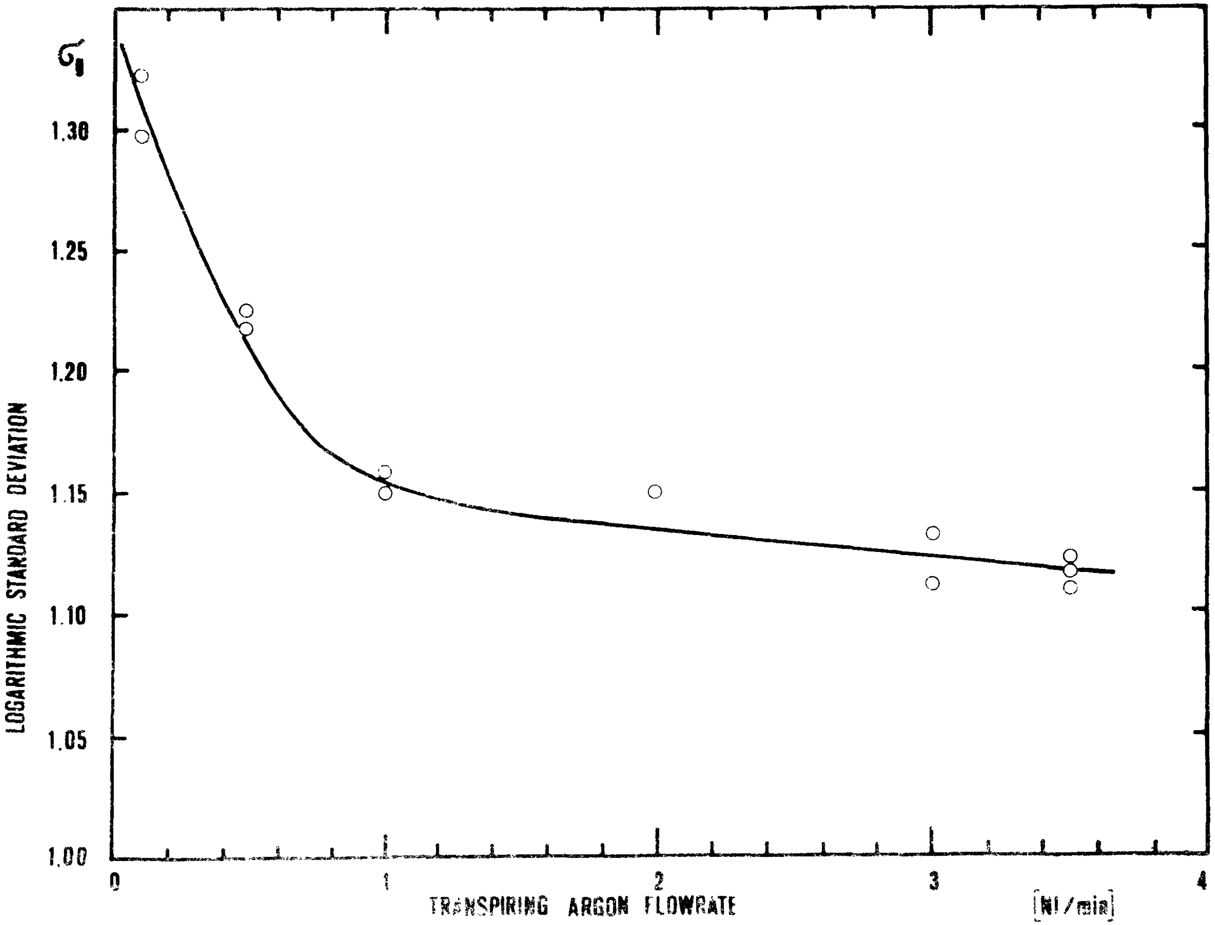

Fig. 6 Logarithmic standard dev. for [illegible] as a function of the argon flowrate transpiring through [illegible]

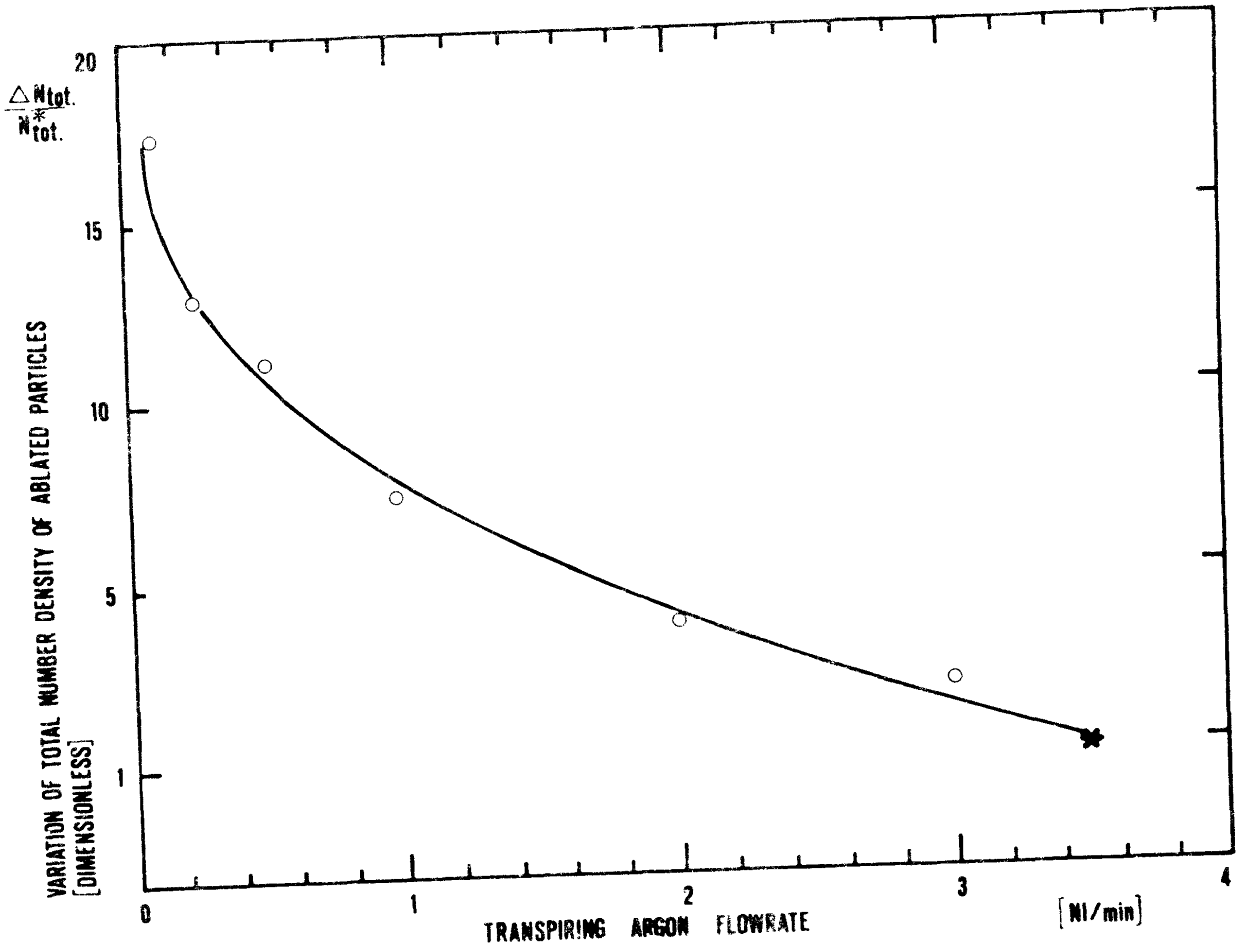

Fig. 7 Variation of total number density of the ablated particles as a function of the argon flowrate transpiring through the porous matrix.

The total concentration values N_{tot}, corresponding to a given mass flowrate, are plotted in dimensionless form according to the following relation:

$$\frac{N_{tot} - N^{*}_{tot}}{N^{*}_{tot}} = \frac{\Delta N_{tot}}{N^{*}_{tot}}$$

where N^{*}_{tot} = 8.736.000 /particle/cm^{3}/ is the reference value and represents the total concentration corresponding to a transpiring argon flowrate of 3.5 Nl/min.

The results given in fig. 7 are in agreement with the qualitative explanation of the aerosol production mechanism given above.

During the experiments consumption of the matrix takes place, and hence, for a given value of the arc current and of the pressure in the plenum chamber feeding the transpiring gas through the matrix, the temperature of the matrix ablating surface decreases. This is due to the lower temperature attained by the ablating surface which can be attributed to two main causes. One is related to the ablation of the matrix which enlarges the inner diametre of the matrix nozzle, decreasing the heat transfer rate from the plasma to the unit area of the nozzle walls. The other one is related to the increase of the transpiring argon flowrate through the matrix.

For a given value of the relative pressure in the chamber and a given matrix temperature distribution the argon flow rate transpiring through the matrix is inversely proportional to the matrix thickness.

The ablation of the matrix reduces its thickness and therefore the mass flowrate increases. This effect is enhanced by the lower temperature attained by the matrix, caused by the decrease in the heat transfer rate from the plasma per unit area of the matrix nozzle, related to the increase in the inner diameter of the ablating nozzle, as above mentioned.

Therefore during a long operation of the particle generator the mean diameter tends to decrease. This can be easily

compensated for by increasing the arc current and decreasing the relative pressure in the plenum chamber for the feeding of the transpiring gas through the matrix.
Further experiments will be performed with arc current up to 300 Amp; various materials will be investigated such as tungsten, ceramic and stainless steel, which are available as porous matrices. Particles from other materials, which are not available as porous matrices, will be generated by mixing the material to be studied with graphite. Since the evaporation temperature of graphite is in most cases higher than that of other elements, it is expected that the admixture predominately evaporates, producing aerosols of the desired material.

BIBLIOGRAPHIC REFERENCES

1) Chace, W.G., Moore, H.K., *Exploding Wires*, Vol.1 (1959), Vol. 2 (1962), Vol.3 (1964), Plenum Press, New York.

2) H. Reichelt, "An Aerosol Generated in Electric Arcs Between Metal Electrodes", Aerosol Research at the First Physics Institute, Wien (1968).

3) J.G. Holmgren, J.O. Gibson, C. Sheer, "Some Characteristics of Arc Vaporized Submicron Particulates", *Ultrafine Particles*, John Wiley & Sons, New York (1963).

4) W.E. Kuhn, "The Formation of Silicon Carbide in the Electric Arc", *Ultrafine Particles*, John Wiley & Sons, New York (1963).

5) Boffa, C., Pfender, E., "Controlled generation of monodisperse aerosols in the submicron range", Journal of Aerosol Science. vol. 4 (1973).

6) Liu, B.Y.H., Whitby, K.T., Pui, D.Y.H., "A portable electrical analyzer for size distribution measurement of submicron aerosols" Journal of the Air Pollution Control Association, vol.24, n.11 (1974).

ISBN 0-12-452950-X